

# Search for broken time-reversal symmetry near the surface of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ films using $\beta$ -NMR

H. Saadaoui,<sup>1,\*</sup> G. D. Morris,<sup>2</sup> Z. Salman,<sup>3,\*</sup> Q. Song,<sup>1</sup> K. H. Chow,<sup>4</sup> M. D. Hossain,<sup>1</sup> C. D. P. Levy,<sup>2</sup> T. J. Parolin,<sup>5</sup> M. R. Pearson,<sup>2</sup> M. Smadella,<sup>1</sup> D. Wang,<sup>1</sup> L. H. Greene,<sup>6</sup> P. J. Hentges,<sup>6</sup> R. F. Kiefl,<sup>1,2,7</sup> and W. A. MacFarlane<sup>5</sup>

<sup>1</sup>*Department of Physics and Astronomy, University of British Columbia, Vancouver, BC, V6T 1Z1, Canada*

<sup>2</sup>*TRIUMF, 4004 Wesbrook Mall, Vancouver, BC, V6T 2A3, Canada*

<sup>3</sup>*Clarendon Laboratory, Department of Physics, Oxford University, Parks Road, Oxford, OX1 3PU, UK*

<sup>4</sup>*Department of Physics, University of Alberta, Edmonton, AB, T6G 2G7, Canada*

<sup>5</sup>*Chemistry Department, University of British Columbia, Vancouver, BC, V6T 1Z1, Canada*

<sup>6</sup>*Department of Physics, University of Illinois at Urbana-Champaign, Urbana, Illinois, 61801, USA*

<sup>7</sup>*Canadian Institute for Advanced Research, Toronto, ON, M5G 1Z8, Canada*

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Weak spontaneous magnetic fields are observed near the surface of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  films using  $\beta$ -detected Nuclear Magnetic Resonance. Below  $T_c$ , the magnetic field distribution in a silver film evaporated onto the superconductor shows additional line broadening, indicating the appearance of small disordered magnetic fields. The line broadening increases linearly with a weak external magnetic field applied parallel to the surface, and is depth-independent up to 45 nm from the  $\text{Ag}/\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  interface. The magnitude of the line broadening extrapolated to zero applied field is less than 0.2 G, and is close to nuclear dipolar broadening in the Ag. This indicates that any fields due to broken time-reversal symmetry are less than 0.2 G.

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The highly unconventional electronic properties of high- $T_c$  superconductors (HTSC) give rise to novel interfacial phenomena that are important fundamentally (e.g. in probing symmetry of the bulk electronic ground state); as well as in applications (e.g. junction-based devices). While significant progress has been made in understanding the transport properties of such interfaces, very little is known about their *magnetic* properties, in part due to the lack of an appropriate local magnetic probe. A particularly unresolved issue is whether the superconducting order parameter (OP) breaks time-reversal symmetry (TRS) near the surface.<sup>1,2</sup> A characteristic feature of TRS-breaking (TRSB) is spontaneous magnetization; however, Meissner screening cancels this in the bulk, limiting the associated fields to within the magnetic penetration depth of defects and interfaces.<sup>3</sup> To measure this magnetization directly, one requires a sensitive depth-dependent local magnetic probe. In this paper we use a novel technique based on depth-controlled beta-detected nuclear magnetic resonance ( $\beta$ -NMR) to search for TRSB order near the surface of the high- $T_c$  cuprate superconductor  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (YBCO).

In contrast to Ru-based and heavy fermion superconductors,<sup>4,5</sup> there is no evidence for TRSB in the bulk of HTSC cuprates, particularly YBCO,<sup>6</sup> where OP-phase-sensitive measurements have established spin-singlet  $d_{x^2-y^2}$ -wave order.<sup>7</sup> There are some indications of weak magnetism,<sup>8,9</sup> some of it related to the  $\text{CuO}$  chains in YBCO,<sup>10</sup> or to vortex cores above the lower critical field  $H_{c1}$ .<sup>11</sup> Surface scattering of the Cooper pairs from most surfaces perpendicular to the  $\text{CuO}_2$  planes frustrates  $d_{x^2-y^2}$ -wave order within a few coherence lengths of the interface.<sup>3</sup> This leaves a high density of mobile holes (as evidenced by the zero bias

conductance peak (ZBCP) in tunneling spectra) that may condense into a superfluid of different symmetry than the bulk,<sup>12</sup> e.g.  $s$ -wave, or TRSB states such as  $d_{x^2-y^2} + is$  and  $d_{x^2-y^2} + id_{xy}$ .<sup>13</sup> Other origins of a TRSB state include, frustrated OP near grain boundaries or junctions,<sup>3</sup> the interaction of a self-induced magnetic field caused by the OP distortion with the OP itself,<sup>14</sup> and finite size effects in thin films.<sup>15,16</sup>

Experiments to detect TRSB near surfaces have yielded controversial results. Carmi *et al.* measured a weak spontaneous magnetic field using SQUID magnetometry near the edges of epitaxial  $c$ -axis oriented YBCO films below  $T_c$ .<sup>17</sup> Spontaneous Zeeman-like splitting of the ZBCP, due to TRSB, has been seen in some tunneling measurements,<sup>18</sup> but not in others.<sup>19</sup> Phase sensitive measurements, which could also detect spontaneous flux, showed no evidence of a TRSB state.<sup>20</sup> A resolution to this disagreement requires more direct information of interface magnetism in cuprates using a local magnetic probe that can locate the origin and distribution of any such fields on the atomic scale.

In this study, we present direct measurements of the magnetic field near the interface of silver and YBCO films using  $\beta$ -NMR. We measure the field distribution using a highly spin polarized  $^8\text{Li}^+$  beam implanted into a thin silver overlayer deposited on YBCO. We find an inhomogeneous broadening of the field distribution below the  $T_c$  of YBCO, with the probe ions stopping at an average distance of 8 nm from the  $\text{Ag}/\text{YBCO}$  interface. By extrapolating the line broadening to zero applied field we find the mean internal field is very close to experimental resolution determined by the Ag nuclear dipolar fields. In this way we obtain an upper limit of 0.2 G for any spontaneous fields of electronic origin.

The experiment was performed using  $\beta$ -NMR of  $^8\text{Li}^+$  at the ISAC facility at TRIUMF in Vancouver, Canada. For details, see Refs.<sup>21,22</sup> Similar to NMR, to measure the resonance, we apply a field along the spin polarization (here in the plane of the films)  $\mathbf{B}_0 = B_0 \hat{y}$  (with  $5 \leq B_0 \leq 150$  G) and follow the polarization of  $^8\text{Li}^+$  as a function of the frequency  $\omega$  of a small transverse radio-frequency (RF) field of amplitude  $B_1 \sim 1$  G, applied along the  $\hat{x}$ -axis. The resonance condition is  $\omega = \gamma B_{\text{loc}}$ , where for  $^8\text{Li}^+$ ,  $\gamma = 0.63015$  kHz/G, and  $B_{\text{loc}}$  is the local field. At this  $\omega$ , the polarization, initially parallel to the  $\hat{y}$ -axis, is averaged by precession in the oscillating field. In the absence of dynamic effects, the resulting resonance is generally broadened by any static inhomogeneity in the local magnetic field. Thus, the lineshape offers a detailed measurement of the distribution of local magnetic fields in the sampled volume determined by the beamspot ( $\sim 3$  mm in diameter) and the implantation profile (see below).

A novel pulsed RF mode was used in this study. The RF field is applied in  $90^\circ$  pulses randomized in frequency order, instead of the continuous wave (cw) mode commonly used.<sup>22</sup> In randomly pulsed RF (RPRF), one obtains a high signal to noise ratio with minimal contribution from both variations in the incoming  $^8\text{Li}^+$  rate and cw power broadening. Because of the limited  $B_1$ , the RPRF mode is suitable for narrow lines up to a few kHz in width. Fig. 1 shows the resonance spectrum at 100 K with a half width at half maximum (HWHM) of approximately  $\Delta_0 \approx 0.15$  kHz (or 0.24 G). Similar cw spectra can be at least twice as broad,<sup>21</sup> making it difficult to resolve a small additional broadening.

Major advantages of  $\beta$ -NMR in detecting TRSB are the abilities (i) to implant the probe  $^8\text{Li}^+$  at low energy into thin layered structures and (ii) to control the mean implantation depth on the nanometer scale. In this study,  $^8\text{Li}^+$  is preferentially implanted into the thin silver overlayer evaporated onto YBCO, instead of the superconductor itself. Stopping the probes in the overlayer eliminates the possibility of the probe perturbing the superconductor. Also, the  $^8\text{Li}^+$  nucleus carries a small electric quadrupole moment, so the spectrum in the Ag is free of any quadrupole splittings which are present in non cubic YBCO.<sup>21</sup> Consequently the resonance of  $^8\text{Li}^+$  in Ag, below 1 Tesla, is single narrow line with a  $T$ -independent linewidth attributed to nuclear dipole broadening from the small nuclear moments of  $^{107}\text{Ag}$  and  $^{109}\text{Ag}$ .<sup>22</sup> From basic magnetostatics, any inhomogeneous fields in the YBCO layer will decay exponentially outside the superconductor as  $\exp(-\frac{2\pi}{a}z)$ , where  $a$  is the length scale of the inhomogeneity.<sup>23-25</sup> Thus we can only detect such fields provided our probe-YBCO stopping distance  $z$  is  $\lesssim \frac{a}{2\pi}$ . Any static field inhomogeneities arising in this way will broaden the intrinsic resonance of the Ag layer.

The measurements presented here were carried out on (110), (103), and (001)-oriented YBCO films capped with 15 or 50 nm of Ag. The (110) film ( $T_c = 86.7$  K) was grown by RF magnetron sputtering on a (110)  $\text{SrTiO}_3$

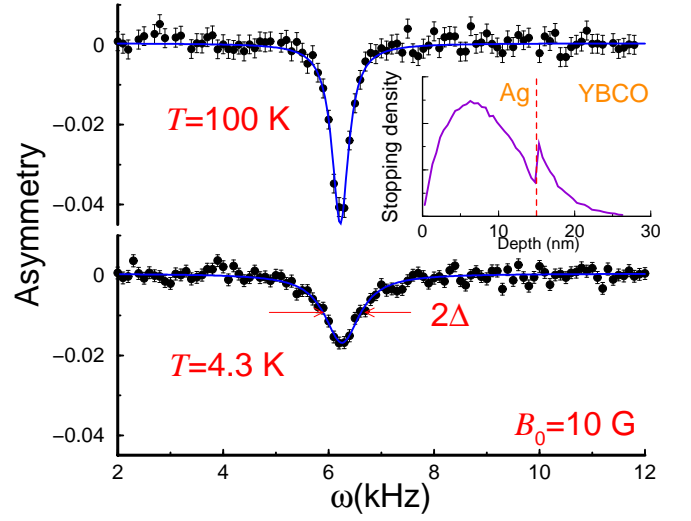


FIG. 1: (Color online). Typical  $\beta$ -NMR spectra taken by implanting 2 keV  $^8\text{Li}^+$  into Ag/YBCO(110), in an external field of  $B_0 = 10$  G (FC) applied along the surface of the film. Solid lines are fits to a Lorentzian of HWHM  $\Delta$ . Inset: simulated implantation profile using TRIM.SP for  $^8\text{Li}^+$  of 2 keV in 15 nm of Ag on YBCO.<sup>26</sup> The  $^8\text{Li}^+$  stops at an average depth of 8 nm away from the Ag/YBCO interface.

(STO) substrate measuring  $8 \times 6$  mm. The (103) film ( $T_c = 84$  K) was grown under similar conditions. Three (001) films were also studied, (i) one with  $T_c = 88.7$  K grown on (001) STO under similar conditions as the (110), and (ii) two films ( $T_c \sim 88.0$  K) grown by thermal co-evaporation on  $8 \times 10$  mm  $\text{LaAlO}_3$ . The films are epitaxial and atomic force microscopy was used to characterize the surface roughness which is in the range 6-12 nm. All samples were capped *ex-situ* with 15 nm of Ag, except one of the last two which was capped with 50 nm, by DC sputtering 99.99% Ag at room temperature under an Ar pressure of 30 mtorr at a rate of  $0.5 \text{ \AA/s}$  while rotating the sample to ensure uniformity. The  $^8\text{Li}^+$  implantation energy was varied so that the probe ions are implanted at average depths ranging from 8 to 43 nm. The inset of Fig. 1, shows the simulated stopping profile of 2 keV  $^8\text{Li}^+$  ions in 15 nm of Ag on YBCO using TRIM.SP.<sup>26</sup> Here the average probe-YBCO distance is  $\sim 8$  nm. At 2 keV, about 20% of  $^8\text{Li}^+$  ions stop in the YBCO, yielding no associated NMR signal due to fast spin-lattice relaxation at low fields. To measure the NMR resonance in Ag, a small field is required, and our measurements were taken by field-cooling (FC) in a small field  $B_0$  or zero field-cooling (ZFC). Residual magnetic fields were reduced to less than 30 mG normal to the surface when FC, or in all directions when ZFC. The samples were aligned parallel to the field with an accuracy of at least  $0.5^\circ$ .

Fig. 1 shows two resonances at 100 K and 4.3 K in Ag/YBCO(110). Above  $T_c$ , the resonances are all identical and show negligible differences in amplitude and linewidth, and are indistinguishable from those intrinsic

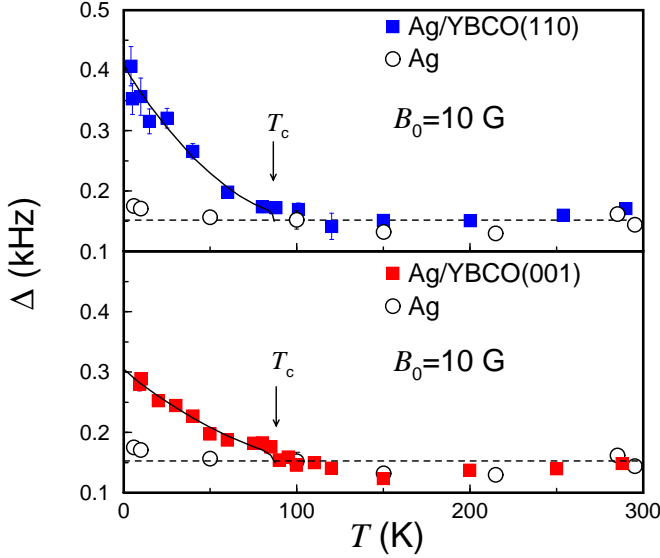


FIG. 2: (Color online). The  $T$ -dependence of the HWHM,  $\Delta$ , of the resonance of 2 keV  $^8\text{Li}^+$  implanted into Ag/YBCO(110), Ag/YBCO(001), and Ag. The data on the Ag/YBCO(110) and Ag film was taken using FC, and ZFC for the Ag/YBCO(001). The widths were independent of FC or ZFC. The dashed lines represent the average Ag width  $\Delta_0$ , the arrows point to the  $T_c$  of the YBCO films, and the solid lines are guide to the eye.

to Ag. Below  $T_c$ , the resonance broadens, therefore is reduced in amplitude. The broadening is symmetric, unlike the field distribution within the bulk of a superconductor in an ordered vortex lattice state.<sup>27</sup> Such a symmetric broadening is typical of a more disordered vortex distribution.<sup>28–30</sup> The HWHM,  $\Delta$ , of a single Lorentzian fit to both (110) and (001) samples is plotted in Fig. 2. It is nearly  $T$ -independent above  $T_c$ , consistent with the small nuclear dipolar broadening in Ag. It is the same in both samples and comparable to a control sample of Ag grown on an insulating STO substrate under similar conditions (open circles, Fig. 2). Below  $T_c$ , the resonance broadens, signaling the appearance of disordered static magnetic fields in the underlying YBCO.

$\Delta$  below  $T_c$  is slightly larger in Ag on the (110) film than the (001) film. In an applied field of 10 G, the additional broadening,  $\Delta - \Delta_0$ , at  $\approx 5$  K is already very small, about 0.25 kHz for the (110) film, and 0.15 kHz for the (001) film. This broadening is clearly caused by the superconducting YBCO, since it is absent above  $T_c$  and in the Ag film without YBCO. It is, however, not accompanied by a resonance shift (see Fig. 1). The resonance frequency is constant from 300 K to 5 K in all films, independent of FC or ZFC cooling. This shows there is no superconducting proximity effect in the Ag layer where an induced Meissner shielding of the applied field leads to a diamagnetic resonance shift, e.g. as seen recently in Ag/Nb heterostructures.<sup>31</sup> The temperature-dependent broadening below  $T_c$  is not linked to TRSB, as the latter state is expected theoretically to appear at a second transition temperature  $T_{c2} \ll T_c$ ,<sup>3,12,13</sup> consis-

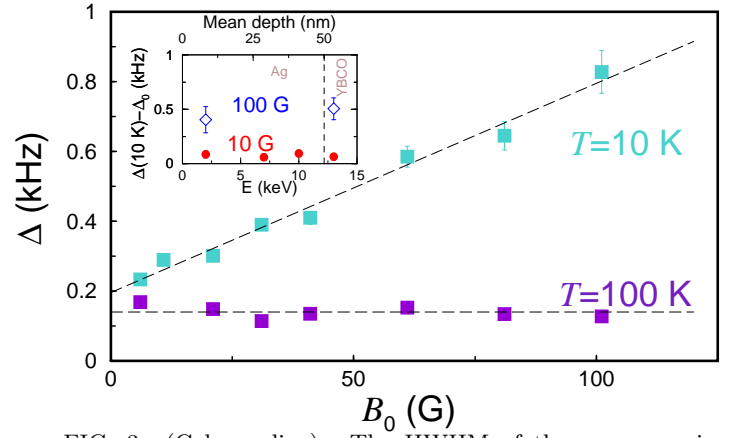


FIG. 3: (Color online). The HWHM of the resonance in Ag/YBCO(001) at 10 K taken after ZFC. The dashed lines are linear fits. Inset: energy and depth dependence of the excess HWHM at 10 K ( $\Delta(10\text{ K}) - \Delta_0$ ) in the 50 nm Ag/YBCO(001) sample at applied fields 10 and 100 G. The resonance at all energies is due to the fraction of  $^8\text{Li}^+$  landing in Ag film.

tent with some tunneling measurements.<sup>18</sup> In contrast, the broadening here has an onset close to  $T_c$ .

The line broadening versus the external magnetic field in the  $c$ -axis sample is displayed in Fig. 3. At 100 K,  $\Delta$  is field-independent as expected. At 10 K (ZFC),  $\Delta$  increases linearly with the applied field. This field-dependent broadening is attributed to inhomogeneous penetration of the applied field in the form of flux vortices. Penetration of vortices would not typically occur at these fields well-below  $H_{c1}$ ,<sup>32</sup> especially since the demagnetization factor for the field parallel to the film is very small. However, at the interface the flux lines may penetrate more easily due to suppression of the  $d$ -wave order near twin or grain boundaries.<sup>33,34</sup> Moreover, surface roughness suppresses the Bean-Livingston surface barrier, and vortices may nucleate at fields  $H \leq H_{c1}$ .<sup>35</sup> Surface vortices have been observed in YBCO crystals in fields as small as 4 G applied parallel to the surface.<sup>36</sup> At such low fields, the vortex spacing  $d$  is of the order of few microns.<sup>37</sup> Outside the superconductor, the resulting field inhomogeneity leads to a depth independent broadening, since  $z \ll d$ , consistent with our results (inset of Fig. 3).

The  $T$ -dependence of the linewidth in Fig. 2 is consistent with line broadening due to vortices beneath the surface since one would expect such vortex penetration close to  $T_c$ , and that it would increase as the temperature falls due to the decreasing magnetic penetration depth in the superconductor.<sup>38,39</sup> Field inhomogeneities are also expected from local variations of the shielding current density due to surface roughness as well as twin and grain boundaries.<sup>40</sup> This inhomogeneity will be enhanced by increasing the field or decreasing the temperature below  $T_c$ ; due to a higher current density with pronounced local variations. This would lead to a temperature and field-dependent broadening as observed in Figs. 2 and 3. It is possible that flux penetration and current density

inhomogeneities near the surface could be related to the apparent dead layer seen in HTSC and other superconductors using low-energy  $\mu$ SR.<sup>41,42</sup> Further experiments on atomically flat surfaces may help elucidate the origin of the magnetic field inhomogeneities reported here. However the field-dependent source of the line broadening is not central to the current study.

The main result of this study is the zero field extrapolation of the broadening at low temperature which is an estimate of the TRSB fields. The broadening at 10 K extrapolates to  $\Delta_{B=0} \approx 0.2$  kHz in the (001) and (103), and 0.3 kHz in (110) (not shown). This  $\Delta_{B=0}$  is marginally higher than the normal state broadening,  $\Delta_0 \approx 0.15$  kHz. Thus, the net internal field in the superconducting state extrapolated to zero applied field,  $\Delta_{B=0} - \Delta_0$  is less than 0.15 kHz (or  $\sim 0.2$  G) in all orientations. Part of the difference is due to the slight broadening of the Ag resonance upon cooling from 100 to 5 K, as seen in Fig. 2 (open circles). Thus, the additional broadening at zero field  $\Delta_{B=0} - \Delta_0$ , is an estimate of the spontaneous magnetic field at the Ag/YBCO interface, and has an upper limit of 0.2 G. This extrapolated value is close to our experimental resolution determined by the Ag nuclear moments. These additional fields at low temperature are clearly much weaker than predicted by tunneling exper-

iments where much stronger spontaneous fields are predicted to cause the ZBCP splitting.<sup>18</sup>

In conclusion, we have conducted a depth-resolved  $\beta$ -NMR study of the field distribution near the interface of Ag and YBCO films. In all films we find additional broadening of the NMR resonance below  $T_c$ , signaling the appearance of disordered internal static fields in YBCO. We established an upper limit of 0.2 G for TRSB fields at low temperature. This rules out any straightforward interpretation based on the TRSB state that was suggested by tunneling measurements to be characterized by a much larger spontaneous magnetic field.<sup>18,19</sup> We have shown that such a putative state must be consistent with a small upper limit on the width of the magnetic field distribution in the adjacent Ag. We have also demonstrated that  $\beta$ -NMR can be used as a sensitive magnetic probe of spontaneous magnetic fields near an interface.

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- \* Present address: Paul Scherrer Institute, Laboratory for Muon Spin Spectroscopy, 5232 Villigen PSI, Switzerland.
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